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# Magnetic force microscopy on cobalt nanocluster films

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## Abstract

Magnetic force microscopy (MFM) has been employed to image the magnetic stray field from cobalt nanocluster films. The uniformly sized clusters with a diameter about 10 nm were deposited at low energy with a gas aggregation source. Both isolated particles supported on silicon, and cluster-assembled films were analyzed. MFM measurements on individual clusters are complicated by interference from surface height variations, while for thicker layers a clear field pattern can be observed. The magnetic correlation length across such films is substantially larger than the size of a single cluster, indicating that the clusters are magnetically coupled to form stable domains. In ex situ magnetization experiments, a small increase or decrease in the MFM phase signal occurred depending on the direction of the applied field.

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**Keywords:** Nanoclusters; Magnetic force microscopy; Magnetic correlation length

## 1. Introduction

Nanostructured ferromagnets are expected to form an important class of materials in future technologies. The properties of isolated magnetic grains change considerably as their sizes enter the nanometer range. Particles smaller than the width of a domain wall (about 40 nm for bulk Fe) are in a single-domain state with the magnetization along the anisotropy axis. Above the onset of superparamagnetism (near 8 nm for a spherical Co grain) such particles have a large coercivity. For applications in magnetic storage media, high anisotropy and low coupling between the grains are required. In contrast, when isolated grains aggregate to form densely packed nanostruc-

tured magnets, the resulting properties are largely determined by the exchange coupling between randomly oriented grains [1]. As a result such materials [2] are often magnetically soft which can make them suitable for the use in transformers for example.

It is important to clarify the dependence of magnetic properties on structural features. For this reason efforts are devoted to image the domain structure in these new materials and their dynamics. In particular, small-angle neutron scattering (SANS) measurements have been employed to characterize magnetic correlation lengths [1,3]. In this paper we explore magnetic force microscopy (MFM) [4,5] as a technique for direct imaging of the stray field pattern arising from Co cluster-assembled films, as well as single nanoclusters supported on a Si substrate. Although MFM can provide useful information about the local magnetic structure, a precise comparison between height and phase images is necessary in order to separate true

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magnetic information from topography interference. Furthermore, the influence of external magnetic fields on the remanent state of Co cluster aggregations is investigated.

## 2. Experiment

Clusters were produced with the NC200U source from Oxford Applied Research, which is a gas aggregation source equipped with a dc magnetron [6]. Co atoms were sputtered with an ion current of 0.26 A into a water cooled condensation/drift region. This region contained Ar at a pressure of  $(2\text{--}5) \times 10^{-3}$  mbar, the base pressure of the source being  $2 \times 10^{-8}$  mbar. Freshly grown clusters were channeled through apertures into a deposition chamber, where Si(1 0 0) wafer substrates at room temperature were exposed for a few minutes up to 45 min to form either open or closed film structures. The final size of the clusters depends mainly on the length of the condensation region; with an intermediate length we obtained clusters about 10 nm in diameter. This size was determined from high-resolution transmission electron micrographs of films with very low coverage (and thus non-aggregated clusters) [7], avoiding also the influence of AFM tip convolution. After exposure to air the Co clusters are covered with an oxide shell of thickness  $\sim 2$  nm. Random stacking of clusters in the low-energy deposition process leads to a porous film morphology, with a density that is expected to be about half of the bulk metal density [8]. The films were sputter-coated (*ex situ*) with 2 nm of Pd prior to imaging, to reduce sticking of clusters to the probe tip.

Imaging was performed in air with the Dimension 3100 AFM from Digital Instruments. The MFM tips were of the commercially available MESP type coated with 50 nm of ferromagnetic CoCr alloy, and magnetized in the *z*-direction parallel to the tip axis. MFM measurements were taken in lift mode, where topography and magnetic field are probed in alternating passes of the tip. During a MFM pass, the tip-to-surface distance is held constant at a predetermined lift height to minimize influence from topography. The tip responds to changes in the magnetic force with a phase shift in the cantilever oscillation [5], where a positive shift results from a repulsive interaction. Finally, external magnetic fields were applied to a Co cluster

film with a thickness of 375 nm. The field of an electromagnet was varied in steps from 2.4 to 11.4 kG, and applied for 1 min in either out-of-plane or in-plane direction, after which remanent (zero field) MFM images were collected.

## 3. Results

### 3.1. Single clusters

Fig. 1 shows the topography of a  $1 \mu\text{m} \times 1 \mu\text{m}$  area of Si substrate with deposited Co clusters, and the corresponding MFM image. The oscillation amplitude in tapping mode was about 22 nm; with a scan height of 12 nm in lift mode this leads to an average tip-to-sample distance of 34 nm. In the topography image the clusters appear to have diameters typically in the range of 30–35 nm, an overestimation that is caused by the increased nominal radius of curvature (25–50 nm) of a magnetic tip. In the MFM image a positive phase shift is observed at each cluster position, with a magnitude ( $\sim 0.5^\circ$ ) that appears to be related to the cluster height. The cluster diameters are identical in both images so that the resemblance between topography and MFM images is very close. A series of measurements was performed where the lift scan height was increased in steps from 12 to 22 nm (Fig. 2), starting from the situation in Fig. 1. The data in the graph correspond to the maximum phase shift on one particular cluster, obtained after applying a low-pass image filter to reduce the influence of noise. A lift height of 12 nm was the minimum required for stable scanning. Above 13 nm there is a fast decrease in the MFM signal, and above 17 nm it is barely higher than the noise level. The latter value corresponds to a tip-to-sample distance of 39 nm. The dependence on lift height followed the same pattern on other Co samples as well.

For verification, the MFM measurements were repeated on samples with deposited niobium (Nb) clusters, giving very similar results. For this material the phase shifts can not be caused by any magnetic interaction. Electrostatic forces could likewise be ruled out by scanning with a non-magnetized tip: this made no difference either on Co or Nb clusters. Very slow scanning at a rate of 0.25 Hz only reduced the noise, while the phase shifts at the clusters remained unchanged.

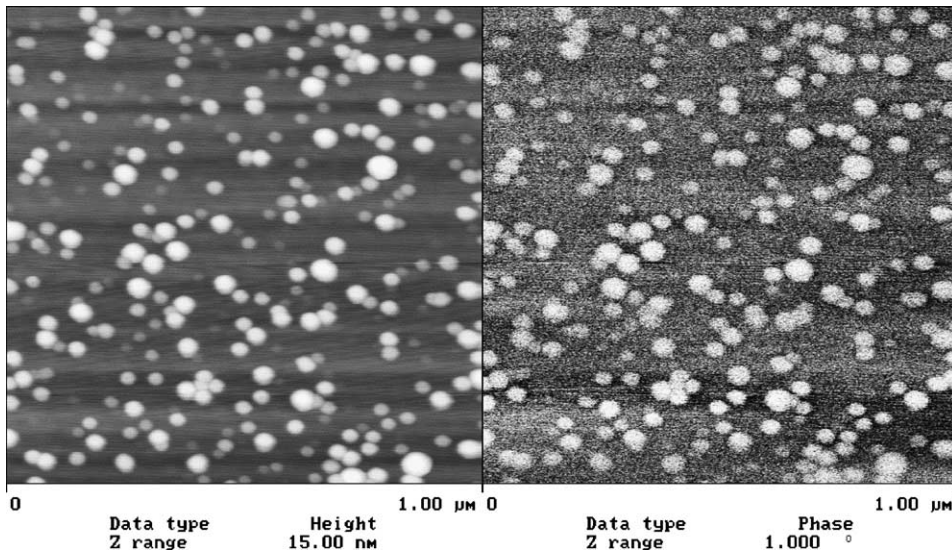


Fig. 1. Height and phase images of Co clusters deposited on Si.

### 3.2. Continuous films

Fig. 3 displays the topography and MFM images of a  $2\mu\text{m} \times 2\mu\text{m}$  area of a Co cluster film with a thickness of 375 nm. An increased lift height of 40 nm was necessary to accommodate the larger fluctuations in surface height. The MFM image is not directly related to the topography. Cluster contours can still be distinguished due to the effect described above, but larger coherent regions are observed where

the contrast is either bright (repulsive interaction) or dark (attractive interaction). These domains extend over numerous clusters, and close examination reveals that the edges of the domains correspond to cluster edges. The rms value of the phase shift over the entire image is  $0.20^\circ$ . Films with smaller thickness down to 75 nm gave comparable results. To quantify the difference in local order, an attempt was made to calculate the correlation lengths for both types of data on  $8\mu\text{m}$  images. The height data exhibit self-affine scal-

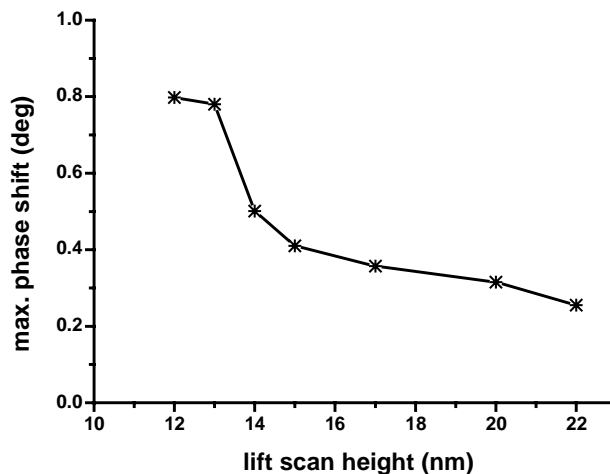


Fig. 2. Highest phase shift detected on a cluster as a function of lift scan height.

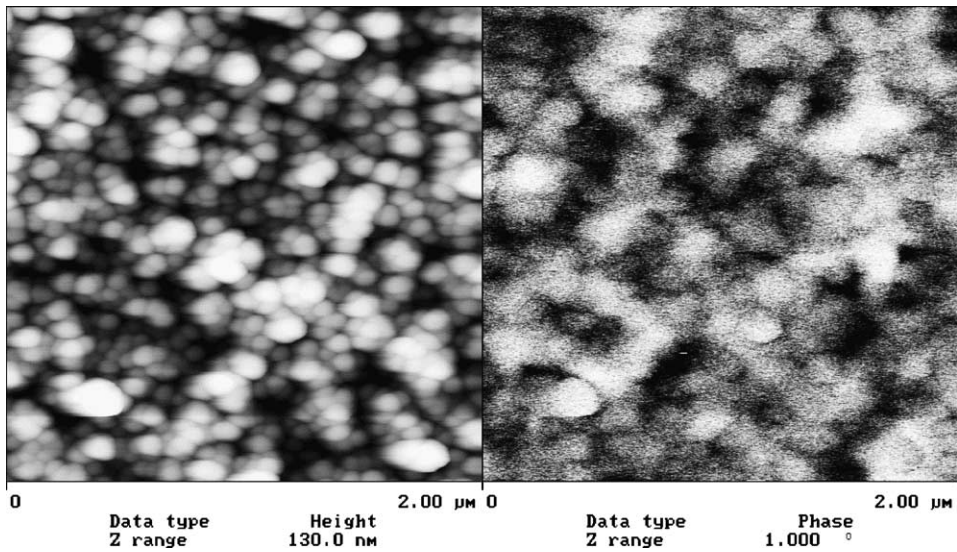


Fig. 3. Height and phase images of a 375 nm thick film of Co clusters.

ing behavior with the height-height correlation function  $g(x) = \langle [h(x) - h(0)]^2 \rangle$  proportional to a power of  $x$  at small lengths  $x$ . At large length scales  $g(x)$  saturates to the value  $2w^2$  with  $w = \{\langle [h(x)]^2 \rangle\}^{1/2}$  the rms roughness amplitude. The correlation length of 91 nm was found from the intersection of power-law and saturation lines in a double-log plot which is close

to the knee regime of the correlation function. However, the phase data do not follow a power-law behavior so that the correlation length (about 200 nm) was estimated directly from the knee regime of the corresponding plot of  $g(x)$  of the phase data.

The 375 nm thick film of Co was exposed to various external fields in an electromagnet, in a sequence

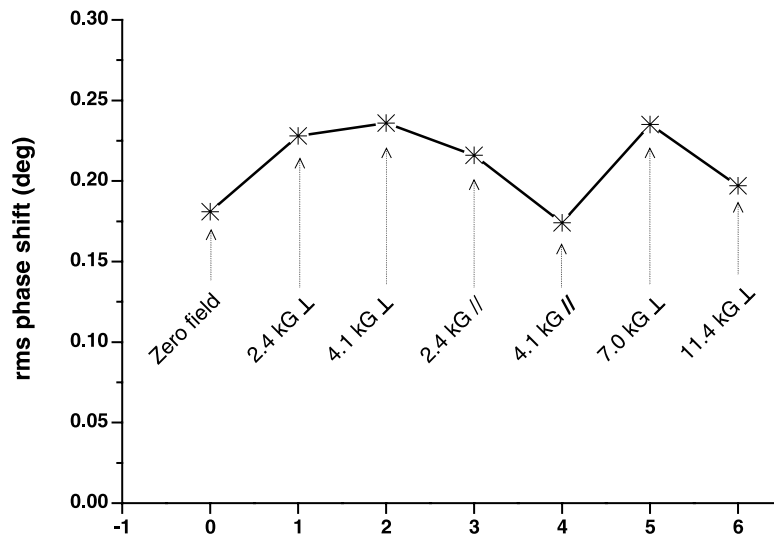


Fig. 4. Root-mean-square values of phase shift over entire MFM images (8  $\mu\text{m}$ ), in remanent state after applying the indicated magnetic fields.

depicted in Fig. 4. Each time MFM images of the remanent state were collected with 2, 5, and 8  $\mu\text{m}$  scan size, where only results from the latter are presented (others were similar). The influence of the external fields is clear from the root-mean-square values of the phase shift in Fig. 4. An increase from 0.20 to 0.29° is observed after the first exposure to 2.4 kG normal to the plane. Increasing the field further to 4.1 kG has little effect, which might indicate saturation. Applying fields parallel to the plane leads to a relatively sharp decrease in signal. This can be expected because MFM is sensitive to second derivatives of the field with respect to the  $z$  direction, at least when the tip is magnetized uniformly along  $z$  and the cantilever is assumed to be parallel to the surface. Finally, with a large field of 7.0 kG normal to the plane the signal increases again, while the subsequent drop at 11.4 kG is unexpected. Changes in the domain structure were not observed.

## 4. Discussion

### 4.1. Single clusters

Experimentally, MFM measurements on single superparamagnetic particles suffer from a commonly observed topographic contamination of magnetic field data [9,10]. It can be suppressed by keeping the lift height large enough during the MFM scan [10,11]. This is in agreement with our findings, where a lift height >17 nm (tip-sample distance  $\sim$ 40 nm) was sufficient to almost eliminate topographic interference. A disadvantage is the loss of sensitivity to weak fields near the surface. The origin of the topographic interference has not been explained in the literature, but a possible explanation is air damping of the cantilever oscillation. Close to the sample the air damping conditions can be significantly changed compared to the freely oscillating cantilever, which might have influence on the measured results [4]. Improvements can be expected from measuring in vacuum to enhance the instrument's sensitivity [9]. In any case, the observed phase shift due to topography is always positive, whereas both superparamagnetic and van der Waals interaction would lead to a negative contrast [9]. Moreover, van der Waals forces only become significant at tip-sample distances below 10 nm [4].

### 4.2. Continuous films

The magnetic structure of Co cluster-assembled films was characterized by estimating the in-plane magnetic correlation length, which generally corresponds to an average size of domains across the sample. The estimated size of  $\sim$ 200 nm is substantially larger than the Co nanoparticles, which indicates that the magnetic structure is not only determined by the deposition of randomly oriented grains but also by the magnetic coupling between those grains. This is in agreement with earlier in situ electrical resistivity and magnetization studies of deposited Co clusters of 6–13 nm diameter, where a transition from superparamagnetic to ferromagnetic behavior was observed at low substrate coverage [12], suggesting that long range dipole interactions between the superparamagnetic clusters exist depending on the coverage. On the other hand, for both Ni and Co porous films consisting of low-energy deposited fcc clusters with a diameter of 3–4 nm, local magnetic order of the extent of a grain was observed [3]. Also in SANS experiments on hcp Co with different grain sizes (10–100 nm), the observed correlation lengths were of the order of the grain size due to the strong magnetocrystalline anisotropy [1]. Only for nanostructured Fe and Ni with grain sizes from 10 to 100 nm the magnetic correlations extended over many grains [1].

## 5. Conclusions

Densely deposited Co nanoclusters form regions of coherent magnetization that are much larger than a single particle. This confirms the existence of long range magnetic interactions between the clusters. Magnetization experiments reveal that such films exhibit weak ferromagnetic behavior. The development of ferromagnetic behavior with increasing substrate coverage could not be observed, because the MFM signal from individual clusters is dominated by topographic interference. The interference signal is only eliminated at large scanning distances between the tip and the sample.

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